

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:
Shinsuke Inoue et al.

Application No.: 10/528,530

Confirmation No.: 8299

Filed: March 18, 2005

Art Unit: 1796

For: NOVEL THERMOPLASTIC POLYIMIDE AND
IMIDE OLIGOMER

Examiner: D. Truong

DECLARATION OF HIROYASU INAGAKI UNDER 37 CFR § 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir or Madam:

I, HIROYASU INAGAKI, hereby declare that:

1. My name is Hiroyasu Inagaki. I am over the age of eighteen years, of sound mind and competent to make this declaration. The facts stated herein are of my personal knowledge, and I know them to be true and correct.
2. I received a Master's Degree in Chemistry from the Graduate School of Science at Hiroshima University in 2002.
3. I am currently employed by Manac Incorporated. I have worked in the field of polyimide chemistry for seven years. Since 2002, I have been involved in the Research Laboratory of polyimide technology.
4. I am a named inventor on the present application, U.S. Patent Application No. 10/528,530.

5. Between October 10-17, 2008, I conducted several experimental tests in which I formed an imide oligomer powder by polymerizing bis(2,3-dicarboxyphenyl)ether dianhydride with bis(4-(4-aminophenoxy)phenyl)sulfone, and using 4-phenylethynylphthalic anhydride as a capping agent, by procedures similar to those described in Example 1 of the present application. To form a comparative imide oligomer powder, I polymerized bis(3,4-dicarboxyphenyl)ether dianhydride with bis(4-(4-aminophenoxy)phenyl)sulfone, and used 4-phenylethynylphthalic anhydride as a capping agent, also using procedures similar to those described in Example 1 of the present application. Thus, the only difference between the two oligomer powders were the acid component used, bis(2,3-dicarboxyphenyl)ether dianhydride (also referred to as 3-ODPA) as compared to bis(3,4-dicarboxyphenyl)ether dianhydride (also referred to as 4-ODPA).
6. Further, between July 10-17, 2009, I conducted additional experimental tests in which I formed an imide oligomer powder by polymerizing 30 mol% of 3-ODPA as an essential acid component and 70mol% of 4-ODPA as an optional acid component with bis(4-(4-aminophenoxy)phenyl)sulfone, and using 4-phenylethynylphthalic anhydride as a capping agent, by procedures similar to those described in Example 1 of the present application. I also formed an imide oligomer powder by polymerizing 50 mol% of 3-ODPA as an essential acid component and 50 mol% of 4-ODPA as an optional acid component with bis(4-(4-aminophenoxy)phenyl)sulfone, and using 4-phenylethynylphthalic anhydride as a capping agent, by procedures similar to those described in Example 1 of the present application. To form an additional comparative imide oligomer powder, I polymerized 10 mol% of 3-ODPA and 90 mol% of 4-ODPA as the optional acid component with bis(4-(4-aminophenoxy)phenyl)sulfone, and used 4-phenylethynylphthalic anhydride as a capping agent, also using procedures similar to those described in Example 1 of the present application. Thus, the only difference among these two oligomer powders was the molar ratio of 3-ODPA to 4-ODPA.
7. Each of the resultant imide oligomer powders was excessively added to several solvents (listed below in Table 1) at room temperature, and then stirred to prepare a suspension. The suspension was filtered, and the filtrate was obtained as "a saturated solution." The saturated solution was measured by weight and then dried over 30 min at 250°C (except

for THF, which was at 100°C), and the residue was measured by weight. Solvent solubility (wt%) was calculated according to the following equation:

$$\text{Solvent solubility (wt\%)} = \text{Dry residue (g)} / \text{Saturated solution (g)} * 100$$

8. Table 1 below shows the obtained solvent solubility results for the imide oligomer powders formed, as described above:

Table 1

Ex. No.	Acid Dianhydride (a molar ratio of 3-ODPA to 4-ODPA)	Diamine component	End-capping Agent	Solvent solubility (wt%)			
				NMP	DMAc	γ -BL	THF
1	4-ODPA (0:100)	BAPS	PEPA	13.0	2.8	0.4	0.2
2	3-ODPA : 4-ODPA (10:90)	BAPS	PEPA	14.0	3.6	1.1	0.29
3	3-ODPA : 4-ODPA (30:70)	BAPS	PEPA	26.0	15.6	5.6	1.0
4	3-ODPA : 4-ODPA (50:50)	BAPS	PEPA	30.8	21.0	11.6	1.4
5	3-ODPA (100:0)	BAPS	PEPA	31.0	35.0	33.0	1.3

Where the following abbreviations are used:

Abbreviation	Chemical Name
3-ODPA	Bis(2,3-dicarboxyphenyl)ether dianhydride
4-ODPA	Bis(3,4-dicarboxyphenyl)ether dianhydride
BAPS	Bis(4-(4-aminophenoxy)phenyl)sulfone
PEPA	4-Phenylethynylphthalic anhydride
NMP	N-methyl-2-pyrrolidone
DMAc	N,N-Dimethylacetamide

γ -BL	γ -Butyrolactone
THF	Tetrahydrofuran

9. The solvent solubility values shown in Table 1 show a drastic difference between imide oligomers formed with 3-ODPA and 4-ODPA. In particular, the polyimide or imide oligomers made with 30 mol% or more of 3-ODPA, such as that claimed by the present invention, have an excellent solvent solubility compared to those using more than 70 mol% 4-ODPA as an acid anhydride.

I further declare that all statements made herein of my own knowledge are true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Respectfully submitted,

Hiroyasu Inagaki
Hiroyasu Inagaki

Date: August 4, 2009